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(54) 【発明の名称】 固体高分子型燃料電池用ステンレス鋼

(57) 【要約】

【課題】 固体高分子型燃料電池において優れた耐食性を発揮するステンレス鋼の提供。

【解決手段】 重量%で、Cr:30%以下を含有し、さらに必要に応じて、Mo:10%以下とNi:25%以下との1種以上を含有し、かつ、これらの成分が $10-0.3 \times ([Cr\%]+3 \times [Mo\%]+0.05 \times [Ni\%]) \leq 5$ を満足し、残部が主にFeからなることを特徴とする固体高分子型燃料電池用ステンレス鋼。

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【特許請求の範囲】

【請求項1】 重量%で、

Cr:30%以下を、

 $10-0.3 \times ([Cr\%] + 3 \times [Mo\%] + 0.05 \times [Ni\%]) \leq 5$

となるよう含有し、残部が主にFeからなることを特徴とする固体高分子型燃料電池用ステンレス鋼。

【請求項2】 重量%で、

Cr:30%以下を、

 $10-0.3 \times ([Cr\%] + 3 \times [Mo\%] + 0.05 \times [Ni\%]) \leq 4$

となるよう含有し、残部が主にFeからなることを特徴とする固体高分子型燃料電池用ステンレス鋼。

【請求項3】 重量%で、

Mo:10%以下と、

Ni:25%以下との1種以上を、さらに含有することを特徴とする請求項1または2に記載の固体高分子型燃料電池用ステンレス鋼。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】電力を直接的駆動源とする自動車、小規模の発電システムなどに用いられる固体高分子型燃料電池に関わる。

【0002】

【従来の技術】近年、自動車用燃料電池の開発が固体高分子材料の開発成功を契機に急速に進展し始めている。固体高分子型燃料電池とは、従来のアルカリ型燃料電池、磷酸型燃料電池、熔融炭酸塩型燃料電池、固体電解質型燃料電池などとは異なり、水素イオン選択透過型の有機物膜を電解質として用いることを特徴とする燃料電池であり、燃料には純水素のほか、アルコール類の改質によって得た水素ガスなどを用い、空気中の酸素との反応を電気化学的に制御することによって電力を取り出すシステムである。

【0003】固体高分子膜は薄くても十分に機能し、電解質が膜中に固定されていることから、電池内の露点を制御してやれば電解質として機能するため、水溶液系電解質や熔融塩系電解質など流動性のある媒体を使う必要がなく、電池自体をコンパクトに単純化して設計できるという特徴がある。

【0004】従来、燃料電池用ステンレス鋼としては、特開平4-247852号公報、特開平4-358044号公報、特開平7-188870号公報、特開平8-165546号公報、特開平8-225892号公報、特開平8-311620号公報にて開示されている高い耐食性が要求される熔融炭酸塩環境で稼動する燃料電池用ステンレス鋼や、また、特開平6-264193号公報、特開平6-293941号公報、特開平9-67672号公報に開示された、数百度の高温で稼動する固体電解質型燃料電池材料の発明がなされてきている。

【0005】しかし、一方、一般に150℃程度(水など使用する冷却媒体の沸点による)までの温度領域で稼

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動する固体高分子型燃料電池の構成材料としては、温度がさほど高くなくことやその環境下で耐食性・耐久性が十分発揮させることが可能であることなどにより炭素系の材料が使用されてきており、このタイプへのステンレス鋼の適用は十分に検討されていない。

【0006】固体高分子型燃料電池の構成材料として炭素を使用する問題点として、コストが高くなることや電池の大きさが大きくなることがあげられており、いずれも固体高分子型燃料電池普及の大きな障害となっているのが現状である。

【0007】

【発明が解決しようとする課題】本発明は、上記のような現状に鑑み、固体高分子型燃料電池のコンパクト化や低コスト化のニーズを満たす炭素材料の代替材料としてステンレス鋼を検討し、使用環境に耐える低コストな成分系を提供することをその目的としている。

【0008】

【課題解決のための手段】本発明者らは、Cr、Mo、Niなどの添加元素をどのような条件で含有させたときに固体高分子型燃料電池用材料として必要かつ十分な性能を発揮できるかを鋭意検討の結果その条件を見出すに至って本発明を完成させたものであって、その要旨とするところは、以下の通りである。

【0009】(1) 重量%で、Cr:30%以下を、 $10-0.3 \times ([Cr\%] + 3 \times [Mo\%] + 0.05 \times [Ni\%]) \leq 5$ となるよう含有し、残部が主にFeからなることを特徴とする固体高分子型燃料電池用ステンレス鋼。(2) 重量%で、Cr:30%以下を、 $10-0.3 \times ([Cr\%] + 3 \times [Mo\%] + 0.05 \times [Ni\%]) \leq 4$ となるよう含有し、残部が主にFeからなることを特徴とする固体高分子型燃料電池用ステンレス鋼。

(3) 重量%で、Mo:10%以下と、Ni:25%以下との1種以上を、さらに含有することを特徴とする請求項1または2に記載の固体高分子型燃料電池用ステンレス鋼。

【0010】

【発明の実施の形態】固体高分子型燃料電池は、水素イオンを選択透過する固体高分子膜を炭素や貴金属の微粒子からなる触媒電極で挟み、それぞれの電極上で起こる水素の酸化反応と酸素の還元反応から電子を取り出すことで電力を発生させる。これらの電子は炭素繊維などの導電体製不織布により構成されるカレントコレクタで収集され、導電性のセパレータへとつながれる。このような基本構造をもつ単セルを直列に積み重ね、全体として必要とされる起電力を発生させる電池とする。

【0011】セパレータ機能としては、上述の電気的導通性のほかに、反応ガスである水素または水素混入ガスと酸素を含有する空気などのガスとが混ざり合わないよう分離する機能や、また、必要に応じて水などの冷却媒体が電池構造の内部を流れるが、冷却媒体と反応ガスと

を分離して循環させる構造的機能が要求される。これまでセパレータなどの固体高分子型燃料電池用部材には主に炭素材料が使用されてきたが、溝切加工などを要する製造にコストがかかるだけでなく、あまり薄くできないので、燃料電池全体の低コスト化とコンパクト化の大きな障害となっていた。そこで、発明者らはステンレス鋼を炭素材料に代替させてこの問題を解決することを想到し、その際に重要な課題の一つとして固体高分子型燃料電池の使用環境に耐える必要十分な添加成分の組み合わせや添加量につき検討した。

【0012】固体高分子燃料電池内を流す燃料となる反応ガスは、純水素、多少の不純物を含有する水素、メタノールなどアルコールや炭化水素の分解ガス（代表組成：25%炭酸ガス、75%水素、数十ppmの一酸化炭素）などであり、他方燃焼を制御する反応ガスは酸素含有ガス、一般には大気中の空気である。固体高分子膜が電解質として機能するためにはある程度の水分が必要で、これらのガスは露点80℃程度に制御される。稼働温度は約90℃が一般的である。

【0013】このような系において、燃料電池は稼働と停止を繰り返すが、まずはセパレータ自身が腐蝕しないことがもっとも重要な点であることは言うまでもない。特にメタノールなどの分解ガスを用いる場合は、その中の炭酸ガスが燃料電池内の結露水などに吸収され酸性溶液となることや固体高分子膜自体が酸性の固体電解質であることなどからセパレータが曝される環境は、常温から水などの冷却媒体の沸点（通常せいぜい150℃程度）までの温度範囲での酸性の水溶液環境となり、pHとしては使用条件によっては2程度まで低下する可能性も指摘されている。一旦腐食され始めると、微量の腐食であってもその腐食部から溶出される金属イオンは固体高分子膜を汚染して水素選択透過機能を阻害することによって電池性能に多大な影響を及ぼす可能性もあるので、腐食は微量イオンを溶出させる程度のものであっても問題となる。

【0014】発明者らは、かかる比較的低温の酸性環境における耐食性に寄与する元素は主にCr、Mo、Niであると予想し、その添加量と組み合わせを変化させたステンレス鋼成分を薄鋼板として試作し、それらを実際にセパレータとして加工して市販の固体高分子膜に白金含有のカーボン微細粒ペーストを塗布・乾燥させたり炭素繊維不織布をカレントコレクタとして燃料電池を構成した。燃料ガスとして水素極側に純水素、あるいは模擬メタノール分解ガス（25%CO₂、75%H₂）を、酸素極側には模擬空気ガス（20%O₂、80%N₂）を大気圧で供給し、電池全体を90℃になるよう高温チャンバー内に保持し、正極から負極に向けて外部に流れる短絡電流の経時変化を測定することによる燃料電池性能の耐久信頼性確認試験（耐久発電試験）を行った。

【0015】なお、この試験に用いた電極部のサイズは100mm×100mmであり、セパレータは板厚4mmの各種ステンレス鋼板にガス流路を切削加工で溝をくりぬいて作成した。試験開始から100日経過した時点で外部電流を測定し、初期の発生電流に対する比率を評価することで耐久信頼性評価の尺度とした。ここではこの比率が0.9を超えれば使用可能であると判断した。電池のサイズ、反応ガス、使用温度など各条件は実用的なものとなるよう十分配慮し、2400時間（100日）連続通電という厳しい条件での試験であるから実用的に使用可能なステンレス鋼を十分に選別可能である。

【0016】上記耐久発電試験の結果、2400時間連続通電後電流/初期電流比が0.9以上であったものについてその成分を整理した結果、Crを必須として、Mo、Niを好ましくは含有した上で、 $10 - 0.3 \times ([Cr\%] + 3 \times [Mo\%] + 0.05 \times [Ni\%])$ （[]は各元素の重量%を表す）にて算出される数値が有効な指標であることを見出した。発明者らの検討の結果、上記式によって算出される数値が5以下であれば純水素を燃料ガスとして使用する場合に十分な特性を示し、さらに、上記式によって算出される数値が4以下であればアルコール類の改質ガスを燃料ガスとして使う場合でも十分な特性を示す。つまり固体高分子型燃料電池のセパレータが曝される環境条件において、ステンレス鋼の耐食発現に関わる基本元素であるCr、Mo、Niの含有量に関する下限界が上記式で表現可能であることを明らかにした点が本発明の最大のポイントである。したがって、かかる指標によって、固体高分子型燃料電池に必要な性能を有するステンレス鋼を特定でき、不要もしくは過大な元素の添加を避けて低コストな材料の提供が可能になる。なお、上記の通りセパレータが曝される環境条件において試験したものであるが、かかる環境が最も厳しい条件であるから他のステンレス鋼製構成部材たとえば積層終端部に用いる終端板などにも適用は十分に可能である。

【0017】本発明においては、純水素を燃料ガスとする環境では、 $10 - 0.3 \times ([Cr\%] + 3 \times [Mo\%] + 0.05 \times [Ni\%]) \leq 5$ 、また、アルコール類の改質ガスを燃料ガスとする環境では、 $10 - 0.3 \times ([Cr\%] + 3 \times [Mo\%] + 0.05 \times [Ni\%]) \leq 4$ を満たすことが重要であり、それぞれの元素の役割などの詳細は必ずしも明らかではないが、以下にそれぞれの添加元素について説明する。

【0018】Crは、本発明が対象とする腐食環境において不動態を形成して耐食性を付与する主要な元素であり単独添加でも効果がある。添加の下限値は上記式の条件に従うようにすることによってその効果を発揮するが、30%を超えて添加しても効果は飽和するので30%を上限とするが、コストを十分に下げるという立場からは23%以下の範囲で上記式を満たすよう調整する。

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【0019】Moは、本発明が対象とする腐食環境において、特に局部腐食を抑制する効果を発揮していると考えられるので添加することが好ましい。添加の下限値は上記式の条件に従うようにすることによってその効果を発揮するが、10%を超えて添加しても効果は飽和するので10%を上限とする。コストを十分に下げるという立場からは7%以下、特に純水素環境では3%以下の範囲で上記式を満たすよう調整する。

【0020】Niは、本発明が対象とする腐食環境においてオーステナイト相を増加させることにより鋼材の耐食性をさらに向上させる効果を発揮していると考えられるので添加することが好ましい。添加の下限値は上記式の条件に従うようにすることによってその効果を発揮するが、25%を超えて添加しても効果は飽和するので25%を上限とする。コストを十分に下げるという立場からは20%以下、特に純水素環境では15%以下の範囲で上記式を満たすよう調整する。

【0021】なお、上記式による規定には関係ないが、耐食性に効果のあるCu：2.5%以下なども、極端なコスト増を伴わなければ適宜添加してもよく、本発明の範囲を逸脱するものではない。また、発明者らの現在までに調査した範囲では、本発明が対象とする環境での耐

食性に対する製造方法の影響はないので、極端な製造欠陥を伴わなければいかなる従来方法で製造したものでも良い。

【0022】

【実施例】実施例として、上記100日の耐久発電試験結果の一例を示し、さらに本発明を詳述する。試験条件などの詳細は上記説明した通りである。表1に挙げた成分を含有するステンレス鋼を試験に供した結果、純水素系環境では、 $10 - 0.3 \times ([Cr\%] + 3 \times [Mo\%] + 0.05 \times [Ni\%]) \leq 5$ を、メタノール改質ガス系環境では、 $10 - 0.3 \times ([Cr\%] + 3 \times [Mo\%] + 0.05 \times [Ni\%]) \leq 4$ なる関係式を満足するステンレス鋼材では100日後の発生電流の経時的低下がわずかであり試験後電流/初期電流比で示した試験成績が0.9以上で、それぞれのガス系環境でのセパレータなどの固体高分子型燃料電池用材料として十分機能することが、逆にそれを外れるものは網掛けにて示したように0.9を下回り十分な機能を有さないことが確認された。

【0023】

【表1】

材料 記号	化学成分(質量%)											式量	100質量%中の炭素量 (質量%)
	C	Si	Mn	P	S	Cr	Mo	Ni	Cu	N			
F1	0.01	0.3	0.12	0.03	0.003	19		0.3	0.41	0.01	4.3	0.83	
F2	0.007	0.14	0.12	0.03	0.007	18.8	1.95			0.002	2.6	0.97	
F3	0.004	0.07	0.07	0.025	0.07	18.75	1.82			0.008	2.7	0.98	
F4	0.01	0.48	0.12	0.025	0.005	21.95	0.83	0.11	0.47	0.01	2.7	0.98	
F5	0.008	0.1	0.13	0.028	0.002	22.3	1.63			0.001	1.8	0.99	
F6	0.07	1.75	0.12	0.021	0.004	13					6.1	0.71	
F7	0.01	0.5	0.35	0.024	0.007	11.03					6.7	0.72	
F8	0.02	0.48	0.53	0.022	0.008	11.85					6.4	0.72	
F9	0.018	0.21	0.85	0.021	0.008	10.85	0.38			0.0075	6.4	0.72	
F10	0.012	0.25	0.82	0.023	0.008	18.3				0.01	5.1	0.83	
F11	0.005	0.1	0.15	0.028	0.003	17.2	0.5			0.01	4.4	0.83	
F12	0.005	0.1	0.09	0.022	0.002	17.2	1.22			0.0088	3.7	0.83	
F13	0.008	0.49	0.24	0.037	0.008	15		0.54			5.5	0.79	
F14	0.023	0.25	0.9	0.031	0.008	11.85					6.4	0.72	
F15	0.16	0.25	0.025	0.025	0.005	13	0	2		0.1	6.1	0.7	
A1	0.04	0.7	1	0.028	0.005	17.5		6.5	2.3	0.3	4.7	0.83	
A2	0.08	0.47	8.43	0.027	0.005	18.18		5.81		0.35	4.5	0.83	
A3	0.08	0.5	11	0.027	0.005	18		6.5		0.35	4.5	0.83	
A4	0.03	0.79	0.55	0.03	0.001	24.8	0.88	12.91		0.0341	1.8	0.99	
A5	0.02	0.5	0.6	0.022	0.001	20.37	3.2	15.31	1.7	0.19	0.8	1	
A6	0.14	0.55	0.57	0.017	0.001	20.2	8.28	17.98	0.87	0.22	-2.0	1	
A7	0.05	0.8	1.92	0.023	0.004	18.45		8.01		0.21	4.3	0.95	
A8	0.08	0.75	1	0.028	0.001	19.07		8.02		0.11	4.2	0.93	
A9	0.014	0.74	1.15	0.024	0.007	18.51		10.27			2.6	0.98	
A10	0.11	0.7	0.8	0.025	0.004	18.8	2.35	16.3			1.1	1	
A11	0.02	0.6	1.38	0.024	0.001	18.49	3.49	13.18			1.9	1	
A12	0.09	1.95	1.89	0.025	0.004	24.2	0.75	13.6	0.74	0.25	4.0	0.91	
A13	0.06	3.49	0.28	0.018	0.003	19.3		13.35		0.1	4.3	0.91	
M1	0.16	0.25	0.25	0.025	0.005	13	2	2		0.13	0.8	1	
D1	0.02	0.48	1.75	0.02	0.001	21.88	2.83	5.15					

式量=10・0.3[(Cr)+3(Mn)+0.06(Ni)]

備考 本材料中の炭素(C) 0.8以下に調整可能

備考 式量=10-0.3(Cr)+3(Mo)+0.06(Ni)
 炭素量=0.8を下回った場合

【0024】

【発明の効果】自動車用発電機や可搬型発電機として有望視されている固体高分子型燃料電池のセパレータなどの材料として最適な成分範囲を特定し、これまでの炭素*40

*に比べ低コストでコンパクト化が可能なステンレス材料の提供が可能となった。したがって、本発明の産業上の価値は極めて高いといえる。

フロントページの続き

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(54) STAINLESS STEEL FOR SOLID HIGH POLYMER TYPE FUEL BATTERY

(57)Abstract:

PROBLEM TO BE SOLVED: To impart excellent corrosion resistance by preparing stainless steel contg. specified ratios of Cr, Mo and Ni, and the balance essential Fe.

SOLUTION: Stainless steel contg., by weight, $\leq 30\%$, desirably $\leq 23\%$ Cr and contg., at need, one or more kinds of $\leq 10\%$, preferably $\geq 7\%$ Mo and $\leq 25\%$, desirably $\leq 20\%$ Ni is prepd. Moreover, in the case pure hydrogen is used as fuel gas, the inequality of $10 - 0.3 \times ([Cr\%] + 3 \times [Mo\%] + 0.05 \times [Ni\%]) \leq 5$ ([] denotes the weight % of each element) is satisfied, and, in the case fuel gas is composed of reformed gas of alcohols, the inequality of $10 - 0.3 \times ([Cr\%] + 3 \times [Mo\%] + 0.05 \times [Ni\%]) \leq 4$ is satisfied. In this way, the stainless steel optimum as the material for a separator of a solid high polymer type fuel battery is obtd. at a low cost.

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CLAIMS

[Claim(s)]

[Claim 1] Stainless steel for polymer electrolyte fuel cells characterized by containing less than [Cr:30%] so that it may be set to $10 - 0.3x([\text{Cr}\%] + 3x[\text{Mo}\%] + 0.05x[\text{nickel}\%]) \leq 5$, and the remainder mainly consisting of Fe by weight %.

[Claim 2] Stainless steel for polymer electrolyte fuel cells characterized by containing less than [Cr:30%] so that it may be set to $10 - 0.3x([\text{Cr}\%] + 3x[\text{Mo}\%] + 0.05x[\text{nickel}\%]) \leq 4$, and the remainder mainly consisting of Fe by weight %.

[Claim 3] Stainless steel for polymer electrolyte fuel cells according to claim 1 or 2 characterized by containing further one or more sorts less than [Mo:10%] and not more than nickel:25% by weight %.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] It is concerned with the polymer electrolyte fuel cell used for the automobile which makes power a direct driving source, a small-scale generation-of-electrical-energy system, etc.

[0002]

[Description of the Prior Art] In recent years, development of an automotive fuel cell is beginning to progress quickly ignited by a development success of solid-state polymeric materials. In the conventional alkaline fuel cells, a phosphoric acid mold fuel cell, a fused carbonate fuel cell, a solid oxide fuel cell, etc., it is the fuel cell which is characterized by using the organic substance film of a hydrogen ion selection transparency mold as an electrolyte unlike a polymer electrolyte fuel cell, and is the system which takes out power by controlling a reaction with the oxygen in air electrochemically using the hydrogen gas obtained by reforming of alcohols besides pure hydrogen to a fuel.

[0003] Since it fully functions, the electrolyte is being fixed in the film, even if thin and a solid-state poly membrane will function as an electrolyte if the dew-point in a cell is controlled, it does not need to use media with a fluidity, such as a water-solution system electrolyte and a fused salt system electrolyte, and has the description that the cell itself can be simplified and designed in a compact.

[0004] Conventionally, invention of the stainless steel for fuel cells which works in the melting carbonate environment where the high corrosion resistance currently indicated in JP,4-247852,A, JP,4-358044,A, JP,7-188870,A, JP,8-165546,A, JP,8-225892,A, and JP,8-311620,A is required as stainless steel for fuel cells, and the solid oxide fuel cell ingredient which works again at the elevated temperature of hundreds of times indicated by JP,6-264193,A, JP,6-293941,A, and JP,9-67672,A has been made.

[0005] However, the ingredient of a carbon system is used by that it is possible for corrosion resistance and endurance to make it demonstrate enough under that temperature is not so high or its environment as a component of the polymer electrolyte fuel cell which generally works on the other hand in the temperature field to about (based on the boiling point of cooling media to be used, such as water) 150 degrees C etc., and application of this type of stainless steel is not fully considered.

[0006] It is raised that the magnitude of that cost becomes high or a cell becomes large as a trouble which uses carbon as a component of a polymer electrolyte fuel cell, and the present condition is that all have been the serious failures of polymer electrolyte fuel cell spread.

[0007]

[Problem(s) to be Solved by the Invention] This invention examines stainless steel in view of the above present condition as alternate material of the carbon material which fills the needs of miniaturization of a polymer electrolyte fuel cell, or low-cost-izing, and sets ***** which offers the low cost component system which bears an operating environment as the purpose.

[0008]

[Means for Solving the Problem] The place which this invention persons make it complete this

invention very much to find out the condition for the ability of the need and sufficient engine performance to be demonstrated as a charge of polymer electrolyte fuel cell material wholeheartedly as a result of examination when alloying elements, such as Cr, Mo, and nickel, are made to contain on what kind of conditions, and is made into the summary is as follows.

[0009] (1) Stainless steel for polymer electrolyte fuel cells characterized by containing less than [Cr:30%] so that it may be set to $10-0.3x([\text{Cr}\%]+3x[\text{Mo}\%]+0.05x[\text{nickel}\%]) \leq 5$, and the remainder mainly consisting of Fe by weight %.

(2) Stainless steel for polymer electrolyte fuel cells characterized by containing less than [Cr:30%] so that it may be set to $10-0.3x([\text{Cr}\%]+3x[\text{Mo}\%]+0.05x[\text{nickel}\%]) \leq 4$, and the remainder mainly consisting of Fe by weight %.

(3) Stainless steel for polymer electrolyte fuel cells according to claim 1 or 2 characterized by containing further one or more sorts less than [Mo:10%] and not more than nickel:25% by weight %.

[0010]

[Embodiment of the Invention] Power is generated by taking out an electron from oxidation reaction of the hydrogen which a polymer electrolyte fuel cell sandwiches the solid-state poly membrane which carries out the selection transparency of the hydrogen ion with the catalyst electrode which consists of a particle of carbon or noble metals, and happens on each electrode, and the reduction reaction of oxygen. These electrons are collected by the current collector constituted with nonwoven fabrics made from a conductor, such as a carbon fiber, and are connected to a conductive separator. A single cel with such basic structure is accumulated on a serial, and it considers as the cell made to generate the electromotive force needed as a whole.

[0011] The function separated so that the hydrogen which is reactant gas besides above-mentioned electric conductivity or hydrogen mixing gas, and gas, such as air containing oxygen, may not be mixed as separator ability, and the structural function to separate and circulate a cooling medium and reactant gas although cooling media, such as water, flow the interior of cell structure again if needed are required. Although the carbon material had so far been used for members for polymer electrolyte fuel cells, such as a separator, mainly, since it could not do not much thinly, cost not only starts the manufacture which requires grooving processing etc., but it had become the serious failure of low-cost-izing of the whole fuel cell, and miniaturization. Then, artificers hit on an idea of making a carbon material substitute for stainless steel, and solving this problem, and examined it per the combination of required sufficient addition component which bears the operating environment of a polymer electrolyte fuel cell as one of the technical problems important in that case, or addition.

[0012] the reactant gas which the reactant gas used as the fuel which passes the inside of a solid-state macromolecule fuel cell is alcohol, such as hydrogen, a methanol, etc. containing the impurity of pure hydrogen some, cracked gas (representation presentation: 25% carbon dioxide gas, 75% hydrogen, dozens of ppm carbon monoxide) of a hydrocarbon, etc., and controls another side combustion — oxygen content gas — generally it is the air in atmospheric air. In order for a solid-state poly membrane to function as an electrolyte, a certain amount of moisture is required, and these gas is controlled at about 80 degrees C of dew-points. Operation temperature has common about 90 degrees C.

[0013] In such a system, although a fuel cell repeats operation and a halt, it cannot be overemphasized that it is the most important point that the separator itself does not corrode first of all. When using especially cracked gas, such as a methanol The environment where a separator is put since the carbon dioxide gas in it being absorbed by the dew condensation water in a fuel cell etc., and serving as an acidic solution and the solid-state poly membrane itself are acid solid electrolytes It becomes an acid water-solution environment in the temperature requirement from ordinary temperature to the boiling point (usually about at most 150 degrees C) of cooling media, such as water, and possibility of falling to about two depending on a service condition as pH is also pointed out. Since the metal ion by which elution is carried out from the corrosion section may have great effect on the cell engine performance by polluting a solid-state poly membrane and checking a hydrogen selection transparency function even if it is the corrosion of a minute amount once it begins to be corroded, even if corrosion is the thing of

extent to which elution of the minute amount ion is carried out, it poses a problem.

[0014] the element comparatively contribute to the corrosion resistance in an acid low-temperature environment which require artificers constituted the fuel cell by make a carbon fiber nonwoven fabric into a current collector, after expect that they be mainly Cr, Mo, and nickel, make the stainless steel component to which the addition and combination be changed as an experiment as sheet steel, actually process they as a separator and make a commercial solid-state poly membrane apply and dry the carbon detailed grain paste of platinum content. as fuel gas — a hydrogen pole side — pure hydrogen or simulation methanolysis gas (25%CO₂ —) Simulation air gas (20%O₂ and 80%N₂) is supplied for H₂ to an oxygen pole side with atmospheric pressure 75%. The whole cell was held in the elevated-temperature chamber so that it might become 90 degrees C, and the durable dependability verification test (durable generation-of-electrical-energy trial) of the fuel cell engine performance by measuring aging of a short-circuit current which flows outside towards a negative electrode from a positive electrode was performed.

[0015] In addition, the size of the polar zone used for this trial was 100mmx100mm, and by cutting, the separator hollowed the slot to the various stainless steel plates of 4mm of board thickness, and created the gas passageway to them. When it had passed since test initiation on the 100th, the external current was measured, and it considered as the scale of durable reliability evaluation by evaluating the ratio to an early generating current. Here, when this ratio exceeded 0.9, it was judged that it was usable. Monograph affairs, such as size of a cell, reactant gas, and service temperature, are enough considered so that it may become practical, and since they are trials on the severe conditions of continuation energization, they can fully sort out usable stainless steel practical for 2400 hours (100 days).

[0016] As a result of the 2400-hour continuation energization aftercurrent / initial current ratio arranging the component about what was 0.9 or more as a result of the above-mentioned durable generation-of-electrical-energy trial, after containing Mo and nickel preferably, having used Cr as indispensable The numeric value computed in $10-0.3x([Cr\%]+3x[Mo\%]+0.05x[nickel\%])$ ([] expresses weight % of each element) found out that it was an effective index. When using pure hydrogen as fuel gas as a result of examination of artificers with [the numeric value computed by the above-mentioned formula] five [or less], sufficient property is shown, and further, even when using the reformed gas of alcohols as fuel gas with [the numeric value computed by the above-mentioned formula] four [or less], sufficient property is shown. That is, in the environmental condition to which the separator of a polymer electrolyte fuel cell is put, the point which showed clearly that the minimum community about the content of Cr, Mo, and nickel which are a basic element in connection with the anticorrosion manifestation of stainless steel can express by the above-mentioned formula is the greatest point of this invention.

Therefore, with this index, the stainless steel which has sufficient engine performance required for a polymer electrolyte fuel cell can be specified, addition of an unnecessary or excessive element is avoided, and offer of a low cost ingredient is attained. In addition, although examined in the environmental condition to which a separator is put as above-mentioned, since these environments are the severest conditions, application is fully possible to the termination plate to be used, other configuration members made from stainless steel, for example, laminating trailer.

[0017] In this invention, in the environment which makes pure hydrogen fuel gas In $10-0.3x([Cr\%]+3x[Mo\%]+0.05x[nickel\%]) \leq 5$ and the environment which makes the reformed gas of alcohols fuel gas Although it is important to fill $10-0.3x([Cr\%]+3x[Mo\%]+0.05x[nickel\%]) \leq 4$ and details, such as a role of each element, are not necessarily clear, each alloying element is explained below.

[0018] Cr(s) are main elements with which this invention forms a passive state in the target corrosive environment, and gives corrosion resistance, and are effective of independent addition. Since effectiveness is saturated even if it adds exceeding 30% although the lower limit of addition demonstrates the effectiveness by making it follow the conditions of the above-mentioned formula, 30% is made into an upper limit, but from the position of fully lowering cost, it adjusts so that the above-mentioned formula may be filled in 23% or less of range.

[0019] As for Mo, it is desirable to add, since this invention is considered to demonstrate the

effectiveness which controls especially localized corrosion in the target corrosive environment. Although the lower limit of addition demonstrates the effectiveness by making it follow the conditions of the above-mentioned formula, even if it adds exceeding 10%, since effectiveness is saturated, it makes 10% an upper limit. From the position of fully lowering cost, it adjusts on 7% or less, especially a pure-water prime ring boundary so that the above-mentioned formula may be filled in 3% or less of range.

[0020] As for nickel, it is desirable to add, since it is thought that the effectiveness of raising the corrosion resistance of steel materials further when this invention makes an austenite phase increasing in the target corrosive environment is demonstrated. Although the lower limit of addition demonstrates the effectiveness by making it follow the conditions of the above-mentioned formula, even if it adds exceeding 25%, since effectiveness is saturated, it makes 25% an upper limit. From the position of fully lowering cost, it adjusts on 20% or less, especially a pure-water prime ring boundary so that the above-mentioned formula may be filled in 15% or less of range.

[0021] In addition, although it is not related to the convention by the above-mentioned formula, less than [Cu:2.5%] which has effectiveness in corrosion resistance, if not accompanied by the increase of extreme cost, you may add suitably, and it does not deviate from the range of this invention. Moreover, in the range investigated by current [of artificers], since the effect of the manufacture approach to the corrosion resistance in the target environment does not have this invention, if not accompanied by the extreme manufacture defect, what was manufactured by what kind of conventional approach may be used.

[0022]

[Example] As an example, an example of the durable generation-of-electrical-energy test result on above-mentioned the 100th is shown, and this invention is explained further in full detail. Details, such as a test condition, are as above-mentioned having given explanation. As a result of presenting a trial with the stainless steel containing the component mentioned to Table 1, it is in a pure hydrogen system environment, $10 - 0.3 \times ([Cr\%] + 3 \times [Mo\%] + 0.05 \times [nickel\%]) \leq 5$ in a methanol reformed gas system environment $10 - 0.3 \times ([Cr\%] + 3 \times [Mo\%] + 0.05 \times [nickel\%]) \leq 4$ — at the stainless steel material which satisfies relational expression, the trial results which the fall of the generating current of 100 days after with time comes out only, there is, and were shown by the trial aftercurrent / initial current ratio or more by 0.9 That in which functioning enough as charges of polymer electrolyte fuel cell material, such as a separator in each gas system environment, separates from it conversely being less than 0.9, as half tone dot meshing showed, and not having sufficient function was checked.

[0023]

[Table 1]

試料 記号	化学成分(質量%)											式値	100日間の貯蔵後試験 純水溶液 (カラム分取) 入系
	C	Si	Mn	P	S	Cr	Mo	Ni	Cu	N			
F1	0.01	0.3	0.12	0.03	0.003	19		0.3	0.41	0.01		4.3	0.93
F2	0.007	0.14	0.12	0.03	0.007	18.8	1.95			0.0092		2.8	0.97
F3	0.004	0.07	0.07	0.025	0.007	18.75	1.82			0.008		2.7	0.98
F4	0.01	0.48	0.12	0.025	0.005	21.95	0.83	0.11	0.47	0.01		2.7	0.98
F5	0.006	0.1	0.13	0.026	0.002	22.3	1.83			0.001		1.8	0.99
F6	0.07	1.75	0.12	0.021	0.004	13						8.1	0.71
F7	0.01	0.5	0.35	0.024	0.007	11.05						8.7	0.77
F8	0.02	0.48	0.53	0.022	0.008	11.85						8.4	0.77
F9	0.018	0.21	0.85	0.021	0.006	10.85	0.98			0.0075		8.4	0.77
F10	0.012	0.25	0.82	0.023	0.008	18.3						5.1	0.85
F11	0.005	0.1	0.15	0.028	0.003	17.2	0.5			0.01		4.4	0.93
F12	0.005	0.1	0.09	0.022	0.002	17.2	1.22			0.0068		3.7	0.95
F13	0.009	0.49	0.24	0.037	0.008	15						5.5	0.78
F14	0.023	0.25	0.9	0.031	0.008	11.85		0.54				6.4	0.65
F15	0.16	0.25	0.025	0.025	0.005	13	0	2		0.1		6.1	0.7
A1	0.04	0.7	1	0.028	0.005	17.5		6.5	2.3			4.7	0.93
A2	0.09	0.47	9.43	0.027	0.005	18.18		5.61		0.3		4.5	0.95
A3	0.09	0.5	11	0.027	0.005	18		6.5		0.35		4.5	0.93
A4	0.03	0.79	0.55	0.03	0.001	24.8	0.88	12.91		0.0341		1.6	0.99
A5	0.02	0.5	0.5	0.022	0.001	20.37	3.2	15.31	1.7	0.19		0.8	1
A6	0.14	0.55	0.57	0.017	0.001	20.2	8.28	17.98	0.87	0.22		-2.0	1
A7	0.05	0.8	1.92	0.023	0.004	18.45		8.01		0.21		4.3	0.95
A8	0.08	0.75	1	0.028	0.001	18.07		8.02		0.11		4.2	0.93
A9	0.014	0.74	1.15	0.024	0.007	18.51		10.27				4.3	0.93
A10	0.11	0.7	0.8	0.025	0.004	18.8	2.33	15.3				2.6	0.98
A11	0.02	0.8	1.38	0.024	0.001	18.48	3.48	13.18				1.1	1
A12	0.09	1.95	1.58	0.025	0.004	24.2	0.75	13.6		0.25		1.9	1
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備考 式値=10-0.3{[Cr]+3[Mo]+0.05[Ni]}
 : 本試験成績が0.9を下回った場合

[0024]

[Effect of the Invention] The component range optimal as ingredients, such as a separator of the polymer electrolyte fuel cell by which promising ** is carried out as the electric organ for automobiles or a portable mold electric organ, was pinpointed, and offer of a miniaturizable stainless steel ingredient was attained by low cost compared with old carbon. Therefore, it can be said that the value on the industry of this invention is very high.

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TECHNICAL FIELD

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PRIOR ART

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(2) Stainless steel for polymer electrolyte fuel cells characterized by containing less than [Cr:30%] so that it may be set to $10 - 0.3x([Cr\%] + 3x [Mo\%] + 0.05x [nickel\%]) \leq 4$, and the remainder mainly consisting of Fe by weight %.

(3) Stainless steel for polymer electrolyte fuel cells according to claim 1 or 2 characterized by containing further one or more sorts less than [Mo:10%] and not more than nickel:25% by weight %.

[0010]

[Embodiment of the Invention] Power is generated by taking out an electron from oxidation reaction of the hydrogen which a polymer electrolyte fuel cell sandwiches the solid-state poly membrane which carries out the selection transparency of the hydrogen ion with the catalyst electrode which consists of a particle of carbon or noble metals, and happens on each electrode, and the reduction reaction of oxygen. These electrons are collected by the current collector constituted with nonwoven fabrics made from a conductor, such as a carbon fiber, and are connected to a conductive separator. A single cel with such basic structure is accumulated on a serial, and it considers as the cell made to generate the electromotive force needed as a whole.

[0011] The function separated so that the hydrogen which is reactant gas besides above-mentioned electric conductivity or hydrogen mixing gas, and gas, such as air containing oxygen, may not be mixed as separator ability, and the structural function to separate and circulate a cooling medium and reactant gas although cooling media, such as water, flow the interior of cell structure again if needed are required. Although the carbon material had so far been used for members for polymer electrolyte fuel cells, such as a separator, mainly, since it could not do not much thinly, cost not only starts the manufacture which requires grooving processing etc., but it had become the serious failure of low-cost-izing of the whole fuel cell, and miniaturization. Then, artificers hit on an idea of making a carbon material substitute for stainless steel, and solving this problem, and examined it per the combination of required sufficient addition component which bears the operating environment of a polymer electrolyte fuel cell as one of the technical problems important in that case, or addition.

[0012] the reactant gas which the reactant gas used as the fuel which passes the inside of a solid-state macromolecule fuel cell is alcohol, such as hydrogen, a methanol, etc. containing the impurity of pure hydrogen some, cracked gas (representation presentation: 25% carbon dioxide gas, 75% hydrogen, dozens of ppm carbon monoxide) of a hydrocarbon, etc., and controls another side combustion -- oxygen content gas -- generally it is the air in atmospheric air. In order for a solid-state poly membrane to function as an electrolyte, a certain amount of moisture is

required, and these gas is controlled at about 80 degrees C of dew-points. Operation temperature has common about 90 degrees C.

[0013] In such a system, although a fuel cell repeats operation and a halt, it cannot be overemphasized that it is the most important point that the separator itself does not corrode first of all. When especially cracked gas, such as a methanol, is used, The environment where a separator is put since the carbon dioxide gas in it being absorbed by the dew condensation water in a fuel cell etc., and serving as an acidic solution and the solid-state poly membrane itself are acid solid electrolytes turns into an acid water-solution environment in the temperature requirement from ordinary temperature to the boiling point (usually about at most 150 degrees C) of cooling media, such as water, and possibility of falling to about two depending on a service condition as pH is also pointed out. Since the metal ion by which elution is carried out from the corrosion section may have great effect on the cell engine performance by polluting a solid-state poly membrane and checking a hydrogen selection transparency function even if it is the corrosion of a minute amount once it begins to be corroded, even if corrosion is the thing of extent to which elution of the minute amount ion is carried out, it poses a problem.

[0014] the element comparatively contribute to the corrosion resistance in an acid low-temperature environment which require artificers constituted the fuel cell by make a carbon fiber nonwoven fabric into a current collector , after expect that they be mainly Cr , Mo , and nickel , make the stainless steel component to which the addition and combination be changed as an experiment as sheet steel , actually process they as a separator and make a commercial solid-state poly membrane apply and dry the carbon detailed grain paste of platinum content . as fuel gas, simulation air gas (20%O₂ and 80%N₂) is supplied for pure hydrogen or simulation methanolysis gas (25%CO₂ and 75%H₂) to an oxygen pole side with atmospheric pressure at a hydrogen pole side, and it becomes 90 degrees C about the whole cell — as It held in the elevated-temperature chamber and the durable dependability verification test (durable generation-of-electrical-energy trial) of the fuel cell engine performance by measuring aging of a short-circuit current which flows outside towards a negative electrode from a positive electrode was performed.

[0015] In addition, the size of the polar zone used for this trial was 100mmx100mm, and by cutting, the separator hollowed the slot to the various stainless steel plates of 4mm of board thickness, and created the gas passageway to them. When it had passed since test initiation on the 100th, the external current was measured, and it considered as the scale of durable reliability evaluation by evaluating the ratio to an early generating current. Here, when this ratio exceeded 0.9, it was judged that it was usable. Monograph affairs, such as size of a cell, reactant gas, and service temperature, are enough considered so that it may become practical, and since they are trials on the severe conditions of continuation energization, they can fully sort out usable stainless steel practical for 2400 hours (100 days).

[0016] Cr is made indispensable as a result of the 2400-hour continuation energization aftercurrent / initial current ratio arranging the component about what was 0.9 or more as a result of the above-mentioned durable generation-of-electrical-energy trial, After containing Mo and nickel preferably, the numeric value computed in $10-0.3x ([Cr\%]+3x [Mo\%]+0.05x [nickel\%])$ ([] expresses weight % of each element) found out that it was an effective index. When using pure hydrogen as fuel gas as a result of examination of artificers with [the numeric value computed by the above-mentioned formula] five [or less], sufficient property is shown, and further, even when using the reformed gas of alcohols as fuel gas with [the numeric value computed by the above-mentioned formula] four [or less], sufficient property is shown. That is, in the environmental condition to which the separator of a polymer electrolyte fuel cell is put, the point which showed clearly that the minimum community about the content of Cr, Mo, and nickel which are a basic element in connection with the anticorrosion manifestation of stainless steel can express by the above-mentioned formula is the greatest point of this invention. Therefore, with this index, the stainless steel which has sufficient engine performance required for a polymer electrolyte fuel cell can be specified, addition of an unnecessary or excessive element is avoided, and offer of a low cost ingredient is attained. In addition, although examined in the environmental condition to which a separator is put as above-mentioned, since these

environments are the severest conditions, application is fully possible to the termination plate to be used, other configuration members made from stainless steel, for example, laminating trailer. [0017] In the environment which makes pure hydrogen fuel gas in this invention Although it is important to fill $10-0.3x([\text{Cr}\%]+3x[\text{Mo}\%]+0.05x[\text{nickel}\%]) \leq 4$ in $10-0.3x([\text{Cr}\%]+3x[\text{Mo}\%]+0.05x[\text{nickel}\%]) \leq 5$ and the environment which makes the reformed gas of alcohols fuel gas and details, such as a role of each element, are not necessarily clear, each alloying element is explained below.

[0018] Cr(s) are main elements with which this invention forms a passive state in the target corrosive environment, and gives corrosion resistance, and are effective of independent addition. Since effectiveness is saturated even if it adds exceeding 30% although the lower limit of addition demonstrates the effectiveness by making it follow the conditions of the above-mentioned formula, 30% is made into an upper limit, but from the position of fully lowering cost, it adjusts so that the above-mentioned formula may be filled in 23% or less of range.

[0019] As for Mo, it is desirable to add, since this invention is considered to demonstrate the effectiveness which controls especially localized corrosion in the target corrosive environment. Although the lower limit of addition demonstrates the effectiveness by making it follow the conditions of the above-mentioned formula, even if it adds exceeding 10%, since effectiveness is saturated, it makes 10% an upper limit. From the position of fully lowering cost, it adjusts on 7% or less, especially a pure-water prime ring boundary so that the above-mentioned formula may be filled in 3% or less of range.

[0020] As for nickel, it is desirable to add, since it is thought that the effectiveness of raising the corrosion resistance of steel materials further when this invention makes an austenite phase increasing in the target corrosive environment is demonstrated. Although the lower limit of addition demonstrates the effectiveness by making it follow the conditions of the above-mentioned formula, even if it adds exceeding 25%, since effectiveness is saturated, it makes 25% an upper limit. From the position of fully lowering cost, it adjusts on 20% or less, especially a pure-water prime ring boundary so that the above-mentioned formula may be filled in 15% or less of range.

[0021] In addition, although it is not related to the convention by the above-mentioned formula, less than [Cu:2.5%] which has effectiveness in corrosion resistance, if not accompanied by the increase of extreme cost, you may add suitably, and it does not deviate from the range of this invention. Moreover, in the range investigated by current [of artificers], since the effect of the manufacture approach to the corrosion resistance in the target environment does not have this invention, if not accompanied by the extreme manufacture defect, what was manufactured by what kind of conventional approach may be used.

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EXAMPLE

[Example] As an example, an example of the durable generation-of-electrical-energy test result on above-mentioned the 100th is shown, and this invention is explained further in full detail. Details, such as a test condition, are as above-mentioned having given explanation. As a result of presenting a trial with the stainless steel containing the component mentioned to Table 1, it is in a pure hydrogen system environment, $10 - 0.3x([\text{Cr}\%] + 3x[\text{Mo}\%] + 0.05x[\text{nickel}\%]) \leq 5$ in a methanol reformed gas system environment $10 - 0.3x([\text{Cr}\%] + 3x[\text{Mo}\%] + 0.05x[\text{nickel}\%]) \leq 4$ — at the stainless steel material which satisfies relational expression, the trial results which the fall of the generating current of 100 days after with time comes out only, there is, and were shown by the trial aftercurrent / initial current ratio or more by 0.9 That in which functioning enough as charges of polymer electrolyte fuel cell material, such as a separator in each gas system environment, separates from it conversely being less than 0.9, as half tone dot meshing showed, and not having sufficient function was checked.

[0023]

[Table 1]

試料 記号	化学成分(質量%)											式値	100日間の耐久保電率 鉛水素系 (カソード付) 入系
	C	Si	Mn	P	S	Cr	Mo	Ni	Cu	N			
F1	0.01	0.3	0.12	0.03	0.003	19		0.3	0.41	0.01		4.3	0.83
F2	0.007	0.14	0.12	0.03	0.007	18.8	1.95			0.0092		2.6	0.87
F3	0.004	0.07	0.07	0.025	0.07	18.75	1.82			0.008		2.7	0.88
F4	0.01	0.48	0.12	0.025	0.005	21.95	0.83	0.11	0.47	0.01		2.7	0.96
F5	0.008	0.1	0.13	0.026	0.002	22.3	1.83			0.001		1.8	0.98
F6	0.07	1.75	0.12	0.021	0.004	13						8.1	0.71
F7	0.01	0.5	0.35	0.024	0.007	11.05						8.7	0.71
F8	0.02	0.48	0.53	0.022	0.008	11.85						8.4	0.71
F9	0.018	0.21	0.85	0.021	0.008	10.85	0.38			0.0075		8.4	0.71
F10	0.012	0.25	0.82	0.023	0.008	18.3						5.1	0.85
F11	0.005	0.1	0.15	0.026	0.003	17.2	0.5			0.01		4.4	0.83
F12	0.005	0.1	0.09	0.022	0.002	17.2	1.22			0.0068		3.7	0.85
F13	0.008	0.49	0.24	0.037	0.008	15						5.5	0.78
F14	0.023	0.25	0.9	0.031	0.008	11.85		0.54				8.4	0.85
F15	0.16	0.25	0.025	0.025	0.005	13	0	2		0.1		6.1	0.7
A1	0.04	0.7	1	0.028	0.005	17.5		6.5	2.3			4.7	0.83
A2	0.09	0.47	9.43	0.027	0.005	18.18		5.61		0.3		4.5	0.85
A3	0.09	0.5	11	0.027	0.005	18		6.5		0.35		4.5	0.83
A4	0.03	0.79	0.55	0.03	0.001	24.8	0.88	12.81		0.0341		1.8	0.99
A5	0.02	0.5	0.5	0.022	0.001	20.37	3.2	15.31	1.7	0.19		0.8	1
A6	0.14	0.55	0.57	0.017	0.001	20.2	6.26	17.98	0.87	0.22		-2.0	1
A7	0.05	0.8	1.92	0.023	0.004	18.45		8.01		0.21		4.3	0.85
A8	0.08	0.75	1	0.028	0.001	19.07		8.02		0.11		4.2	0.83
A9	0.014	0.74	1.15	0.024	0.007	18.51		10.27				4.3	0.83
A10	0.11	0.7	0.8	0.025	0.004	19.8	2.35	15.3				2.8	0.99
A11	0.02	0.8	1.38	0.024	0.001	18.49	3.49	13.18				1.1	1
A12	0.09	1.95	1.59	0.025	0.004	24.2	0.75	13.6		0.25		1.9	1
A13	0.06	3.49	0.28	0.018	0.003	19.3		13.35	0.74			4.0	0.91
M1	0.16	0.25	0.25	0.025	0.005	13	2	2		0.1		4.3	0.81
D1	0.02	0.48	1.75	0.02	0.001	21.68	2.93	5.15		0.13		0.8	0.99

備考 式値=10-0.3{[Cr]+3[Mo]+0.05[Ni]}
 : 本試験成績が0.8を下回った場合

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